

Thin Film Superconducting Devices

J. E. Mercereau and H. A. Notarys

California Institute of Technology, Pasadena, California 91109
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Techniques have been developed with which it is possible to fabricate superconducting thin film structures ("bridges") which show Josephson-like phenomena, with a wide variety of electrical and superconducting parameters. These bridges—based on the proximity effect—are made in layered thin film substrates which have been fabricated from many different, both hard and soft, superconducting materials. The fabrication techniques and the electrical and superconducting characteristics for these proximity effect bridges including a simple low frequency (≤ 10 GHz) equivalent circuit will be discussed. These bridges have been incorporated into simple thin film circuits for use as galvanometers, magnetometers, gradiometers, detector arrays, etc. Extension of these techniques to more complex superconducting thin film bridge circuits including resistors, capacitors, and inductors will be indicated.

INTRODUCTION

In 1962 Josephson¹ predicted that the current produced by electron tunneling between two superconductors, separated by a thin insulating barrier, would have certain unusual characteristics. In particular, he predicted that this current would oscillate at a frequency which depends linearly on the voltage maintained across the barrier, and the phase of this oscillation would be determined by the magnetic field within the barrier. The relationship between frequency ν and voltage V contained only the fundamental constants e , electron charge, and h , Planck's constant, such that $\nu = 2eV/h$ and led to Josephson's current relation

$$j = j_J \sin \left[(2e/h) \int V dt + \alpha \right],$$

where α is associated with the magnetic field in the barrier. In the decade since Josephson's prediction, these effects have been confirmed in every detail and studies of the Josephson effect are a large part of present day research in superconductivity. In fact, these phenomena are sufficiently well understood and technically under control that the Josephson voltage-frequency relationship has been recently adopted by the U. S. Bureau of Standards as the official means by which to maintain the U. S. standard volt.

Josephson's original prediction involved a tunneling process, and superconducting tunnel junctions showing these quantum effects, composed of two superconductors separated by a very thin insulating barrier, are called Josephson junctions. However, it has turned out that the tunneling process is not a necessary requirement for producing the Josephson effects. It has been found that the Josephson phenomena occur between any pair of "weakly connected" superconductors. Precisely what constitutes "weakly connected" is still somewhat obscure from a fundamental point of view, but a number of techniques have been developed in various laboratories by which to fabricate such superconducting junctions. A start has been made to characterize these junctions in terms of their electronic properties and to use them in more complex circuits and devices. This paper

will outline a particular technique for weakly coupling two superconductors and will indicate some of the characteristics of this type of superconducting junction or "bridge." We will emphasize the fabrication techniques for these bridges and some of the more complex circuits which have been developed and give only brief reference to the underlying physics of these particular circuits. However, since these devices employ a superconductor rather than an insulator as a coupling medium, some modification of the original Josephson analysis is necessary and we attempt to outline physically how it comes about.

WEAKLY CONNECTED SUPERCONDUCTORS

Many of the characteristics of superconductivity can be explained on the basis that superconductivity represents a macroscopic quantum state. By that we mean that a single wave function $\Psi = \psi e^{i\varphi}$ can be used to describe the collective behavior of large numbers of superconducting electrons. The number density of the co-operating electrons is given by $\rho = \psi^2$. In this discussion we will be mostly concerned with the supercurrent that this state can carry. Electric current density in this description comes from the basic quantum mechanical definition of current in terms of the velocity operator as

$$j = \text{Re} \left(\Psi^* \frac{e\hbar}{im} \nabla \Psi \right). \quad (1)$$

Performing the indicated operations on the wave function Ψ leads to a current in terms of the amplitude ψ and gradient of the phase φ :

$$j = -\frac{e\hbar}{m} \psi^2 \nabla \varphi. \quad (2)$$

This description implies a direct connection between the quantum mechanical phase φ and experimentally observable parameters such as the supercurrent, and was proposed by London before 1935.

By weakly connecting two superconductors we mean that there is some small region of space in which the amplitude ψ of the wave function is small compared to

its amplitude in the surrounding superconductors. Such a situation is illustrated in the top of Fig. 1, where we have plotted the amplitude of the superconducting wave function as a function of position through a region W connecting superconductor S_1 to superconductor S_2 . This figure illustrates an equilibrium situation in which a single wave function of spatially varying amplitude can be used to describe the superconductivity. In this situation an expression for the supercurrent can be obtained from Eq. (2), inserting the proper values for the amplitude and the gradient of the phase at each point. The current so described will be a true supercurrent, that is, a current which can flow even in the absence of any voltages.

However, there is a maximum current, or critical current, which can be achieved in this situation. When this critical current is exceeded a voltage is developed in the superconductor and we have a regime which is probably best described as nonequilibrium superconductivity. That situation can be indicated as shown in the bottom half of Fig. 1. The superconducting wave function is now composed of two parts: one, Ψ_1 , associated with superconductor S_1 , and the other, Ψ_2 , associated with superconductor S_2 . In the connecting region W these two wave functions overlap and to a first approximation superconductivity in the connecting region W can be described as a two phase state $\Psi_w = \Psi_1 + \Psi_2$. The electric current in this situation can still be calculated in terms of the basic definition of current given in Eq. (1). When this is done, it turns out that the appropriate expression for supercurrent density from superconductor 1 to superconductor 2 is given by the following²:

$$j = j_J \sin(\varphi_1 - \varphi_2) + B \nabla \varphi \{1 + \cos(\varphi_1 - \varphi_2)\}. \quad (3)$$

The first part of expression (3) is just Josephson's original expression for the tunneling supercurrent. The phase difference $(\varphi_1 - \varphi_2)$ is related to the voltage V by

$$(\varphi_1 - \varphi_2) = \frac{2e}{\hbar} \int V dt + \alpha.$$

The second term in (3) also arises from quantum interference effects but appears only in situations of very high current density such as occur in the junctions which we are considering. It should be noted that under a constant applied voltage the time average of Josephson tunnel current is zero. Thus there is no net current in response to a constant voltage and the dissipation is zero. However, the time average of the second term can be finite, indicating that it arises from a basically nonequilibrium characteristic of superconductivity. The basic expression, Eq. (3), can be collapsed into an empirical relationship as follows:

$$j = J_1 + J_2 \cos\left(\frac{2e}{\hbar} \int V dt + \beta\right),$$

where the empirical parameters J_1 , J_2 , and β can be evaluated in terms of the fundamental parameters j_J , B , $\nabla \varphi$, and α .

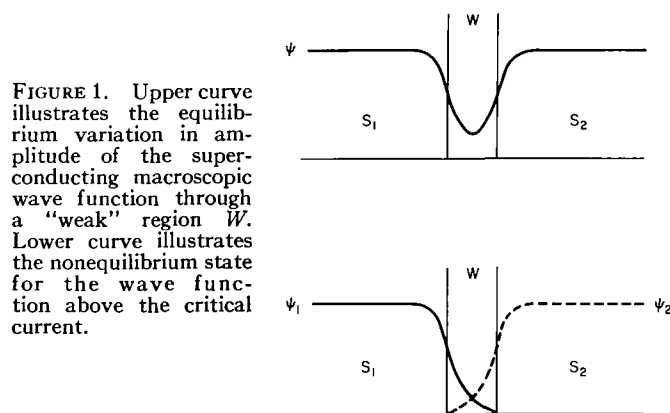


FIGURE 1. Upper curve illustrates the equilibrium variation in amplitude of the superconducting macroscopic wave function through a "weak" region W . Lower curve illustrates the nonequilibrium state for the wave function above the critical current.

For these junctions we find experimentally that this expression can be simplified further and an appropriate expression for the total supercurrent in these junctions at a finite voltage V is given by

$$I_s = \frac{1}{2} I_c \left(1 + \cos \frac{2e}{\hbar} \int V dt\right),$$

where I_c is the temperature dependent critical current for the junction as described previously.

Since these junctions operate in a basically nonequilibrium, dissipating mode of superconductivity, we have found it convenient to describe the operation of the junction in terms of a dissipation voltage V , which is developed across the junction by an impressed current I . These junctions are of such low impedance that in almost all situations they can be considered as being driven from a current source. The form of the dissipation voltage is

$$V = IR - \frac{RI_c}{2} \left(1 + \cos \frac{2e}{\hbar} \int V dt\right), \quad (4)$$

where R is the resistance of the junction measured in its normal state and I_c is the critical current. This simple form for the voltage-current relationship for these junctions has been found to hold for a wide variety of materials and dimensions and involves only measured parameters.

From the form of expression (4) it is evident that an equivalent circuit for this type of junction is a voltage source whose amplitude is v_s , in series with a resistance R . The amplitude of the voltage source is $v_s = RI_c/2$ and its frequency is determined by the voltage V developed across the junction. This voltage source is not a source of power, however, and simply indicates that for a given current the dissipation in the superconducting state is less than in the normal state but that the dissipation is time dependent. Expression (4) is actually an experimental result which has come out of measurements³ on a large number of junctions of various dimensions, resistances, critical currents, and material combinations.

Figure 2 illustrates a typical measured current-voltage characteristic for one of these junctions and this characteristic is accurately predicted by the time

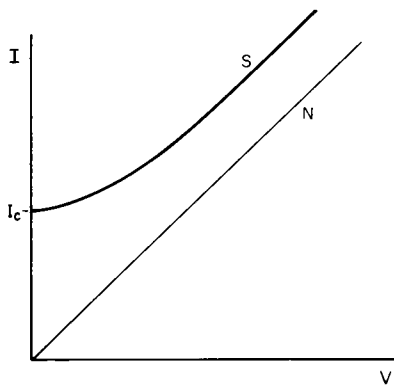


FIGURE 2. Typical current-voltage curve for these junctions. The curve N represents the junction in its normal state, where $V=IR$; curve S represents the junction in its superconducting state. The equation represents the time dependence of the voltage in the superconducting state.

$$V = RI - \frac{RI_c}{2} \left(1 + \cos \frac{2e}{\hbar} \int V dt \right)$$

average behavior of expression (4). The time dependent components of the voltage developed across these junctions have also been measured³ and they too are accurately represented by Eq. (4). Thus we feel at least this type of junction is probably best characterized as a parametric resistance where the resistance is time dependent at a frequency which is related to the voltage developed across it. Notice that at a given voltage these superconducting junctions actually dissipate more power in the superconducting state than in the normal state. This additional dissipation comes about because of the nonequilibrium nature of the superconducting process and arises because of the irreversible periodic destruction of superconductivity within the weak section. This particular form for the equivalent circuit for this type of junction has been verified over a wide range of materials, currents, and dimensions by direct measurements³ of the ac and dc components of the voltage. The equivalent circuit has also been used to analyze⁴ the response of more complex circuits containing these junctions with quite satisfactory results.

FABRICATION TECHNIQUES

In the last few years we have developed general techniques for the fabrication of superconducting thin film structures containing well-defined inhomogeneities in the superconducting wave function, whose electrical and superconducting parameters can be easily controlled. The Josephson-like characteristics of these structures have been investigated over a wide range of electrical and material parameters. The techniques are based on the use of the proximity effect⁵ to locally vary the relative transition temperature (and hence the amplitude or "strength" of the superconducting wave function) between various parts of a composite superconducting film.

When a normal film is superimposed directly on a superconducting film with no intervening oxide layer (or vice versa) the transition temperature of the resultant superconducting sandwich is depressed below that of the superconductor itself. In the thin film limit the transition temperature of the sandwich depends on the relative thicknesses of the normal and superconducting

films and decreases as the normal material thickness is increased, for a fixed superconductor thickness. A similar situation exists for a sandwich of two superconductors of different transition temperature. This composite thin film material we call a superconducting thin film "substrate." Since the transition temperature is a measure of the relative "strength" of the superconductivity, as the transition temperature decreases the "weaker" the superconducting sandwich film becomes. Hence by locally varying the relative thicknesses of superimposed normal and superconducting films, it is possible to develop rapid variations in the amplitude of the superconducting wave function within the thin film substrate.

In this way a thin film structure of the form shown in Fig. 3 can be constructed where the inhomogeneous region of length l' , width w' , and thickness $(t_n' + t_s')$ has a characteristic transition temperature T_c' lower than the surrounding T_c of the main film. This will result in a spatial variation of the wave function such as shown in Fig. 1. But the transition temperature of the inhomogeneous region of length l' will be T_c' only if l' is sufficiently large. For sufficiently short l' the transition temperature of the inhomogeneous region will be higher than its characteristic transition temperature T_c' owing to proximity effects from the adjoining two films. This results from the same proximity phenomenon which determines T_c of the substrate but is caused by a different geometrical aspect of the film and may include interference between proximity effects from the two adjoining films. Owing to this proximity effect across the inhomogeneous region the T_c' can be very low but the actual transition temperature, as determined by the onset of a supercurrent, may be close to that of the adjoining films.

It is thus possible to vary the coupling strength between the main superconducting film sections by varying T_c' (i.e., t_n' , t_s') and the length l' . For a given T_c' there is a range of l' over which Josephson-like effects are generated. However, if l' is too short the coupling between the two superconducting film sections becomes too strong and the film acts as a continuous homogeneous superconductor; conversely, if l' is too long the coupling is destroyed by fluctuations and the film acts as three separate but electrically connected superconductors. As T_c' decreases the necessary l' for Josephson-like effects becomes shorter. The parameters T_c' and l' are detailed functions of the particular materials and films and need to be determined experimentally in order to maximize the Josephson-like effects for a particular material combination.

We have fabricated these thin film structures from many different superconducting materials and find that the Josephson characteristics are in general independent of material. However, because of specific properties of the different materials the fabrication techniques used fall into two general classes: one for soft superconductors and the other for hard superconductors. We will treat only the hard superconducting films here.⁶

The hard superconductor structures were fabricated using Nb, Ta as the superconductor and Ta, W, and Zr as the underlying normal metal. There was only slight alloying between these films which occurred during film fabrication and this could be neglected. The primary cause for the variation of transition temperature in these structures was via the proximity effect as previously described. However, very thin, hard superconducting films themselves, have transition temperatures which vary somewhat with thickness,⁷ probably because of the variation of the structure of the film as it grows. Hence, when two superconducting films with different transition temperature are superimposed, both the proximity effect and the inherent thickness variation of T_c are available as possible modes of local weakening of superconductivity.

The proximity effect junction that has been most extensively studied is composed of Nb on Ta thin film structures. We describe here the fabrication of the Nb/Ta structure as an example of the use of a hard superconductor substrate. The Nb/Ta structure is as shown in Fig. 3(b). First 200 Å Ta is evaporated on a heated sapphire wafer immediately followed by deposition of 100 Å Nb. The superposed film structure has a transition temperature of around 6 K. In general the hard superconducting films were produced by electron beam evaporation onto a sapphire substrate. During evaporation the pressure was typically between 10^{-7} and 10^{-8} mm and the sapphire substrate was maintained at 400°C. This superimposed film is coated with photoresist, and a narrow line is exposed across the film and developed away to uncover a narrow line of film material. A drop of electrolyte is applied over this exposed line of film; then contact to the electrolyte is made by an immersed gold electrode. A voltage pulse is applied between the film as the anode and the gold cathode in order to decrease the thickness of the narrow exposed line of film material by anodization. By varying the applied voltage and pulse duration it is routinely possible to control the depth of anodization (i.e., the film

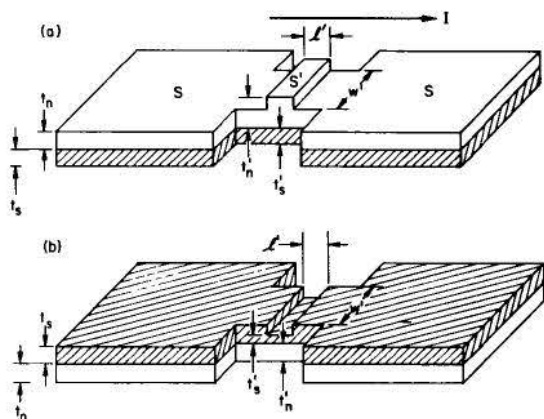


FIGURE 3. Artist's representation of inhomogeneous thin film structures constructed in layered films with superconducting film thickness t_s and normal film thickness t_n . The inhomogeneous area has length l' along the current direction, width w , and thickness ($t_n' + t_s'$). (a) Variable normal film thickness; (b) variable superconducting film thickness.

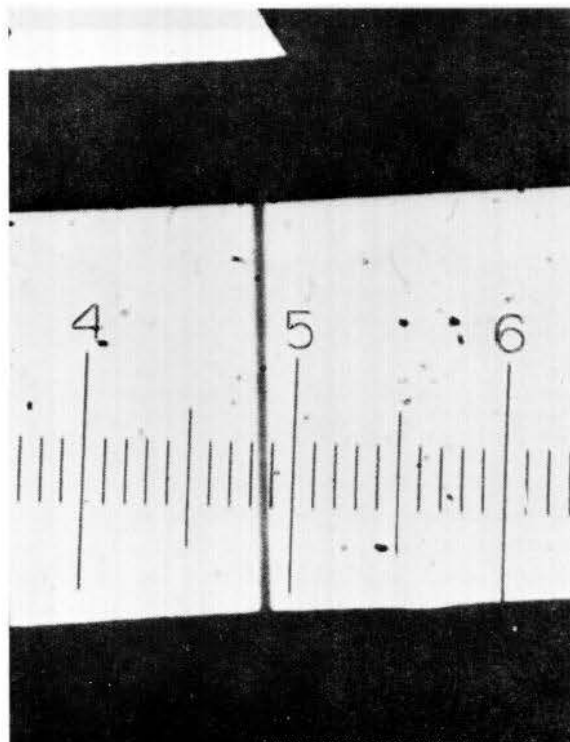


FIGURE 4. Reflection microscope picture of a hard superconductor bridge with scale 1.2μ /division. Main film (light area in picture) is 100 Å Nb on 200 Å Ta and has a transition temperature about 6 K. Bridge region (dark line separating light main film areas) is 50 Å thick with T_c' approximately 2 K. Direction of current flow is through the narrow dimension of the bridge.

thickness) and also control the undercutting. Then with these same photoresist techniques the width of the structure at the exposed line can be made to any desired size by anodization completely through the film. Indium contacts pressed onto the film complete the thin film structure.

The range of parameters over which these structures have been observed to show Josephson-like effects are length 0.3–5 μ , widths 0.5–300 μ , inhomogeneity thicknesses 50–250 Å, T_c' from 4.2 down to below 1.3 K, and normal resistances 10^{-2} –50 Ω . These film properties, lengths, widths, thicknesses, and resistances are typical ranges for all the hard superconductors. Because of the particular properties of these materials, the good film to substrate bond, the hard, protective, and easily controllable oxide, and the fine grain nature of the films, it is possible to reproducibly work with well-defined dimensions down to at least 0.3 μ . The anodization process which is extremely well controlled is crucial to obtain this resolution. These structures are much more stable than the soft superconductors and although lifetime tests have not been completed, circuits are now operating which show no change in characteristics over a period of a year.

JUNCTIONS AND CIRCUITS

A microphotograph of a typical junction is shown in Fig. 4. The horizontal light strip is the superconducting film, crossed by a dark line which is the anodized weak section. The dimensional scale is 1.2 μ per small divi-

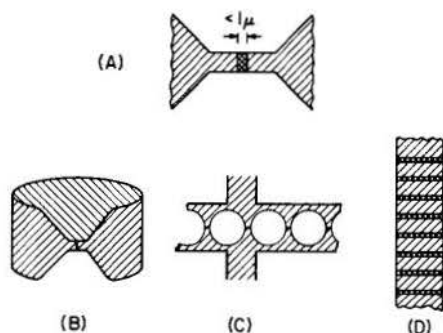


FIGURE 5. Artist's conception of several superconducting film structures. (a) Single bridge in film; (b) single bridge in a cylindrical ring; (c) bridges in parallel (quantum interferometer); (d) bridges in series (corrugated superconductor).

sion, which implies that the length of this junction in the direction of the current flow is less than $\frac{1}{2} \mu$. Voltage developed across this weak section for an impressed current I is described by Eq. (4). If $I \gg I_c$, then the dc voltage \bar{V} is approximately $\bar{V} = R(I - I_c/2)$, and there is also an approximately sinusoidal voltage of frequency $\nu = 2e\bar{V}/h$ and amplitude $RI_c/2$. At lower currents, where $I \gtrsim I_c$, the time average voltage is much less than that just mentioned and the time dependent voltage becomes pulselike with a repetition rate $\tau = h/2e\bar{V}$.

These junctions have been incorporated into more complex superconducting circuits, as indicated by the artist's conception in Fig. 5. Figure 5(a) is the single junction imbedded in a superconducting film such as we have been discussing. Figure 5(b) illustrates a junction in a superconducting cylindrical ring. This type of circuit is the basis for superconducting thin film magnetometers and gradiometers.^{4,8} These devices have a typical magnetic field sensitivity of better than 10^{-10} G with a 1 sec response time and are in routine use in our laboratory for magnetochemical analysis of biological material. Although Fig. 5(b) illustrates a cylindrical film ring, such circuits also operate quite satisfactorily in the form of planar film rings.⁹

Figures 5(c) and 5(d) illustrate other configurations which have been tried. Figure 5(c) is a multiple junction quantum interferometer.¹⁰ As many as 13 junctions have been assembled in parallel in this manner, leading to multiple quantum interference effects as would be expected from such configurations. Figure 5(d) illustrates a number of junctions in series.¹¹ This particular arrangement of "corrugated superconductivity" is being studied with regard to the interactions between the junctions both via the electromagnetic fields and, internally, via the quantum wave function. This configuration may have some applications as a detector of high frequency radiation. Figure 6 is a microphotograph of a typical array of many junctions in series which has been studied in our laboratory. This array was made by a photographic process involving successive exposures for the separate lines (or weak regions). As many as 20 junctions in series have been examined by this technique. We are now developing a procedure to adapt laser interferometry to simultaneously expose large arrays of

lines.¹⁴ By this technique a laser interferogram will be used to expose the photoresist and produce a regular array of uniform lines caused by the constructive and destructive interference of the laser light. Exposures of this type will generate thousands of lines and the usefulness of the technique will ultimately depend on the homogeneity of the superconducting films.

Figure 7 is a composite diagram illustrating a superconducting flux coupled galvanometer along with a microphotograph of the superconducting galvanometer section.¹² It is included as an example of a more complicated superconducting circuit which has been designed and tested to explore the usefulness of these techniques and circuit analysis. This particular device has a current sensitivity of about 10^{-9} A and a zero input impedance, and will probably be used in conjunction with an array of corrugated superconductors as a radiation detector.

These techniques of fabrication are not confined to the investigation of hard superconducting bridges or combinations of bridges. Because of the particular properties of the materials used, the fabrication of entire superconducting microcircuits is possible, including the bridges, resistors, capacitors, and simple inductors. This discussion has centered about the use of a two material composite superconducting film substrate for the fabrication of bridges showing Josephson-like effects. However, the composite superconducting film substrate has more general properties which can be extended to basic studies of superconductivity as well as to microcircuit technology.

The superconducting substrate need not be confined to two-layered materials but can be extended to several

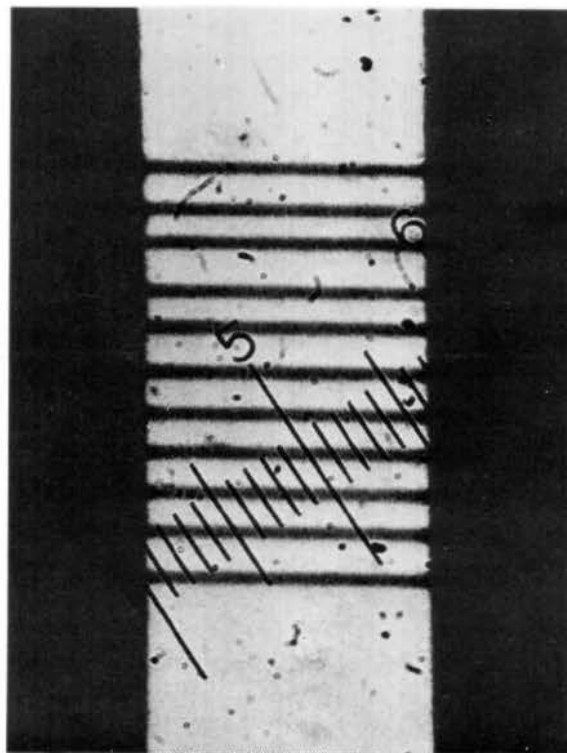


FIGURE 6. Reflection microphotograph of several junctions arranged in series. Scale is 1.2μ per small division.

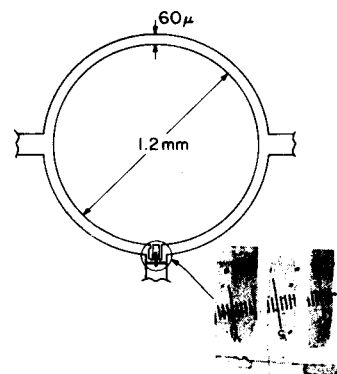
layers with different transition temperatures. Structures of up to five layers have been tested. Depending on the order that these films are deposited the variation of the transition temperature with film thickness may be arranged to increase monotonically, decrease monotonically, or oscillate. If the transition temperature of the film substrate decreases as the thickness decreases and the bottom component film is normal, both superconducting and normal components can be introduced into the microcircuit. The oxides of the hard superconductors lend themselves to the fabrication of capacitors by subsequent evaporation, while simple inductors can also be made by anodizing appropriate film configurations. Furthermore the materials that can be used in this application provide a large range of working temperatures. The elements Hf, Zr, Ti, Nb, Ta, and W cover the temperature range below 9 K. However, other compounds of these elements and high temperature materials such as NbN and NbAlGe can extend this range almost to 20 K and well into the available refrigerator temperature range.

In summary, we have developed techniques for fabricating stable, multilayered thin film superconducting substrates which can then be experimentally manipulated to locally modulate the strength of superconductivity, hence forming an inhomogeneous superconductor. This type of inhomogeneous superconductivity not only leads to Josephson effect devices, multiple arrays of these devices, and combinations of these devices with standard components into an entire microcircuitry, but also provides a form of superconductivity, whose precise behavior is of great interest in its own right.

The fabrication techniques are extremely general and any method for removing or altering microscopic regions of the substrate is applicable. It should be emphasized that it is the spatial modulation of the wave function which is important in this application. These substrates have been developed so that experimentally controllable changes in the thickness of the film results in a controllable spatial modulation of the wave function. In contrast with other forms of "weak link"¹³ it is not the smallness of dimension itself which is critical here. In fact, for many of our configurations the weak section is actually thicker than the surrounding material.

Up to now we have mainly used anodization in conjunction with photoresist techniques. But other means

FIGURE 7. Thin film flux coupled superconducting galvanometer. Insert shows a microphotograph of the superconducting galvanometer section.



such as ion beam etching and ion implantation in conjunction with optical and electron beam resist techniques are also possibilities, and, in particular, ion beam etching has already been successfully used.¹¹ These additional techniques will also extend the usable materials for long term stable substrates out of the limited refractory group used with anodization. Hence an extremely versatile superconducting system has been developed for both microcircuit technology and fundamental investigations of superconductivity.

ACKNOWLEDGMENT

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